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The Cs-137 contamination of soils in the Netherlands and its consequences for the contamination of crop products

M. J. Frissel, J. F. Stoutjesdijk, A. C. Koolwijk and H. W. Köster

National Institute of Public Health and Environmental Hygiene (RIVM), P.O. Box 1, NL 3720 BA Bilthoven, Netherlands

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Abstract

On 2-4 May 1986 the Dutch territory was contaminated with radioactive cesium originating from the nuclear accident at Chernobyl. Radiation measurements indicated that the major part of the contamination was associated with the rainfall. A sampling and analysis programme confirmed this assumption. To estimate the uptake of Cs-134 and Cs-137 by crops in the future use was made of the data of the Soil-to-Plant Transfer working group of the IUR (International Union of Radioecologists). The IUR data provide predictor values for the transfer of radionuclides for particular types of crops and soils. Correction factors are presented for the impact of pH, organic matter content and reduction of the availability of radionuclides with time. Uncertainties associated with averaging time and space effects and local differences as well as 95% confidence limits are provided. The predicted levels for edible parts of crops in the Netherlands vary between 0.1 and 10 Bq kg^{-1} on a dry weight base. On a fresh weight base these values are even lower. The predicted transfer values are compared with results of uptake experiments at RIVM. It can be concluded that, for the range of conditions tested, there is no indication of severe deviations from the predicted uptake due to local conditions in the Netherlands. Therefore the derived equations for the prediction of the uptake of Cs-137 can be applied.

Introduction

On 2 and 3 May 1986, a radioactive cloud, which originated from the nuclear reactor accident in Chernobyl, passed the Netherlands from south-east to north-west. The radiation level during the passage increased at Bilthoven from its normal level of 5-6 microröntgen (μR) per hour within a few hours to almost 10 $\mu R\ h^{-1}$, but decreased in the afternoon of 3 May to about 8 $\mu R\ h^{-1}$ indicating that relatively low amounts of radioactivity had been deposited on the soil surface. In the evening of 3 May some local rain showers developed which contaminated the soil surface considerably as appeared from an increase of the radiation level to 15 μR h^{-1}. During the first hours of 4 May, at ground level, the radioactive cloud seemed to have disappeared. More rain showers followed, once again contaminating the soil. It became clear that cold, uncontaminated air, which had moved in from the west, was already present at ground level. However, the warmer contaminated air was still present in the higher layers of the atmosphere. At the boundary between cold and warm air showers developed which washed considerable amounts of radioactivity from the air. The distribution of rain over the Netherlands was very uneven. Fig. 1 shows that the intense rain showers covered only a small band with a width of a few tens of kilometres. This prompted RIVM to investigate the Cs deposition distribution over the Netherlands.

Contamination of the soil

Sampling and analysis

Soils were sampled to a depth of 5 cm, in a grid of 20 km by 20 km. Per sampling site 3 to 4 subsamples were taken. Total weight per sample was about 1 kg. If possible, sites were selected on flat permanent grasslands. Arable land and sites with sandy soils with a very low organic matter content were avoided. The samples were analysed by gamma-spectrometry with a germanium-lithium detector.

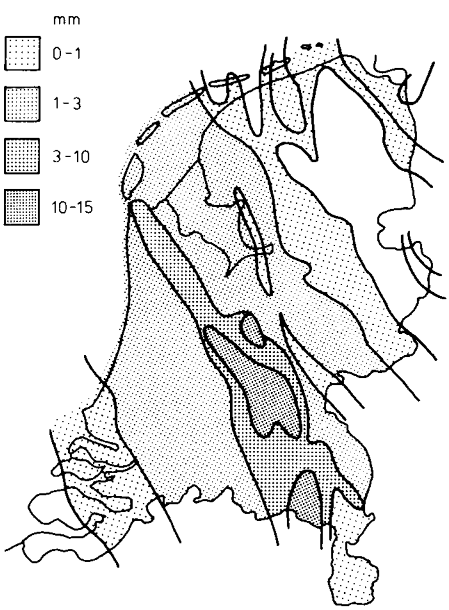

Fig. 1. The rainfall in the Netherlands between 3 May 1986 08h00 and 4 May 1986 08h00 (KNMI data).

The contamination level

The main long-lived contamination consists of Cs-134 and Cs-137. The former one originates exclusively from the Chernobyl accident, whereas of the latter one a considerable part originates from the fall-out of the atomic bombs tested in the atmosphere between 1954 and 1982. Therefore the Cs-134 distribution is selected for presentation (Fig. 2). The Chernobyl Cs-137 levels in Western Europe were on the average two times as high as the Cs-134 levels. Also in the Netherlands this was observed, although in some areas a higher Cs-137/Cs-134 ratio was measured. This ratio has, however, not been investigated further because of other priorities. An overall picture of the Cs-137 deposition level is shown in Table 1. Because Cs-137

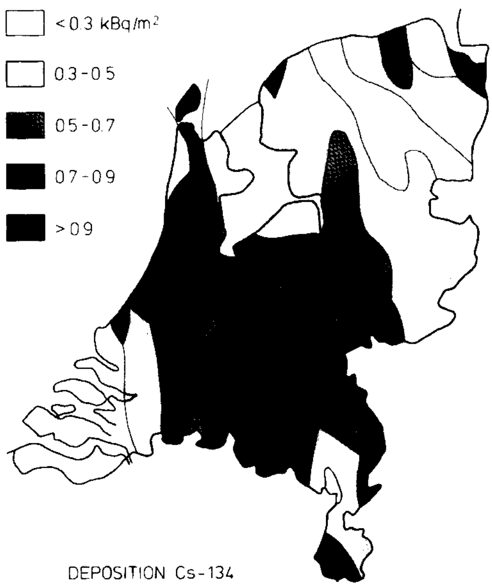

Fig. 2. The Cs-134 distribution in the Netherlands measured during the summer of 1986. Sampling grid distance 20 km × 20 km (RIVM data).

Table 1. Cs-137 deposition levels in the Netherlands

	Deposition level ($Bq\ m^{-2}$)	Distribution
Total 1954-1982	4600	even
Remainder of the 1954-1982 deposition in soils in 1987 (Köster et al., 1987)	1500	variable
Estimated dry, May 1986	500	even
Estimated wet, May 1986	0-5500	variable

has a half-life of 30.2 years compared to 2.1 years of Cs-134, in the remaining part of this paper only Cs-137 will be mentioned. Of course, there is no difference in radioecological behaviour of Cs-134 and Cs-137.

The soil-to-plant transfer factor

Cesium is an element which is closely related to potassium and many phenomena which can be observed for K are observed for Cs too. There are many soils which fix K by means of an almost irreversible adsorption on exchange sites of particular clay minerals. For the clay mineral illite this occurs because after adsorption of K at interlayer positions the mineral or crystal lattice closes. So, Sumner & Bolt (1962), using K-42, found under circumstances which prevail in nature that part of the K-ions which are situated in the interlayer positions of illite exchange against other ions very slowly indeed.

This phenomenon has also been observed for Cs-137. As the amount of Cs-137 in a soil is very small as compared to ever present clay minerals such as illite, practically all soils fix Cs-137 quite well. Also soils with very high percentages of organic matter such as peat soils and many soils in the Nordic countries fix Cs-137, but its availability for the roots of crops seems to be higher than in illite-containing soil. Addition of K enhances the desorption of Cs (Frissel et al., 1981).

Both Cs and K are readily taken up from the soil by the roots of the vegetation and transferred to other parts of the plants. The contamination of a crop is often described by a concentration ratio, usually called transfer factor (TF), which is defined by:

$$TF = Bq\ kg^{-1}\ \text{product (dry weight)}/Bq\ kg^{-1}\ \text{dry soil} \quad (1)$$

Sometimes TF is defined per kg of fresh crop product. In particular for food crops this seems convenient. The disadvantage is that the uncertainties associated with the determination of TF values increase by using the fresh weight based equation. For animal feed the dry weight convention is used exclusively; in this publication this convention is used for all TF values. Despite chemical similarities between Cs and K the TF values for Cs are two orders of magnitude lower than for K (Frissel & Koster, 1987). Studies on the uptake of radiocontaminants are as old as the atomic bomb tests. However, it were the requests for risk analysis calculations of nuclear facilities which in fact started the present interest in the topic. There is an important difference between the study of the uptake of nutrient elements and radionuclides. The soil-plant relation of K is focused on the amount of fertilizers needed for optimum yield. The aim of the study of the uptake of Cs is to derive TF values for the prediction of the contamination of the crop product. They have to consider the total amount of radioactive Cs which may contaminate the soil. A soil analysis method which determines the Cs-137 status of a soil in relation to its plant uptake, as exists for K, has therefore not been developed. TF values for different radionuclides differ considerably, but even for one particular radionuclide, such as Cs-137, the range is large. TF values differ per crop and depend on the type of soil, pH, soil organic matter content and many other variables. The availability of several radionuclides decreases with time, this is particularly true for Cs. Diffusion of Cs-ions into the soil mineral matrix, followed by incorporation, is seen as the explanation of this phenomenon.

The wide variation occurring between experimentally determined TF values prompted the IUR (International Union of Radioecologists) some years ago to initiate a joint project of various radioecological research institutions. The aims of the joint project are twofold: first, to provide guidelines which have to be followed in order to obtain reliable TF values; and second, to determine TF values of selected crops (intercomparison of participating institutions) as well as of a wide variety of crops (for extrapolation to non-investigated conditions).

IUR working group Soil-to-Plant Transfer Factors

The activities of the IUR related to the soil-to-plant transfer are covered by a special working group. In order to be able to guarantee the reliability of the transfer factors, recommendations for uptake experiments have been developed. They have to be followed by members of the working group. In this way manipulations which lead to extremely high or extremely low transfer factors can be avoided. Recommendations exist for several experimental conditions. Important are the ones which specify minimum dimensions and weights for lysimeters and pots and the ones which ensure equilibrium between radiocontaminants in soil solutions and soil matrixes. About 12 institutions in 8 countries participate in the programme for the determination of Cs-137 uptake data.

Result of data collection

So far the IUR has collected about 3600 TF values. All data are stored into a data bank together with relevant information on the labelling and on the environmental conditions of the site on which the crops were grown. By means of statistical analysis prediction values have been estimated. An example is Eq. 2 to be used for all crops, with the exception of grass for which Eq. 3 should be used (Frissel & Koster, 1987). TF_{stand} is the value for a particular radionuclide, crop and soil under standard conditions. TF_{actual} is the TF value under actual conditions, which can be derived from TF_{stand} by applying Eq. 2 for all crops, except grass for which Eq. 3 should be used. The standard conditions imply a pH of 6, a soil organic matter percentage (OM) of 4 for all crops except for grass where 10 is used. Furthermore the labelling depth (Depth, cm) is 20 cm and there is no irrigation (irr, mm), while the period that the radionuclide is present in the soil is set at 2 years (Period, years).

$$TF_{actual} = TF_{stand} \cdot e^{-0.64\,(pH-6)} \cdot e^{0.11(OM-4)} \cdot e^{-0.017(Depth-20)} \cdot e^{-0.082(Period-2)} \cdot e^{0.008(irr)} \qquad (2)$$

$$TF_{actual} = TF_{stand} \cdot e^{-0.64\,(pH-6)} \cdot e^{0.11(OM-10)} \cdot e^{-0.017(Depth-10)} \cdot e^{-0.082(Period-2)} \cdot e^{0.008(irr)} \qquad (3)$$

Standardized transfer values TF_{stand} are presented in Table 2. A peat soil should be considered as a sandy soil with a soil organic matter content of 10%. For soil organic matter percentages higher than 10, a value of 10 should be used.

Table 2. Standardized TF values[a] for Cs-137 and Cs-134 for different products (edible parts) and soils. (Bq kg^{-1} dry weight of food or animal feed per Bq kg^{-1} dry soil.)

Crop[b] type	Expected values (lower limit[c]-upper limit[c])		
	sand	clay	loam
Cereals	0.018 (0.014-0.022)	0.015 (0.011-0.020)	0.014 (0.011-0.018)
Fodder	0.060 (0.044-0.081)	0.050 (0.034-0.074)	0.048 (0.035-0.066)
Grass	0.14 (0.11 -0.18)	0.12 (0.085-0.16)	0.11 (0.091-0.14)
Pods	0.10 (0.068-0.15)	0.084 (0.053-0.13)	0.081 (0.054-0.12)
Root crops	0.063 (0.040-0.098)	0.053 (0.031-0.098)	0.051 (0.032-0.080)
Tubers (potatoes)	0.089 (0.066-0.12)	0.074 (0.051-0.11)	0.072 (0.053-0.096)
Vegetables	0.22 (0.17 -0.29)	0.19 (0.13 -0.27)	0.18 (0.14 -0.24)

[a] All data refer to standard conditions, i.e. a contamination period of 2 years, a pH of 6 and a soil organic matter content of 4% for all crops except for grass where a soil organic matter content of 10% is used.
[b] Crops with similar transfer behaviour have been grouped together in the following 7 different crop types:
Cereals = grain of barley, wheat, oats, rye, maize
Fodder = maize (without grain), clover, lucerne
Grass = grass, vegetation of pastures and grassland
Pods = pods and seeds of peas and beans
Roots = roots of root crops such as beet and radish
Tubers = potatoes, swedes
Vegetables = green vegetables such as lettuce, cabbage, spinach, leek.
[c] The upper and lower limits are the limits of the 95% confidence interval.

Statistical analysis

The statistical analysis to estimate the prediction values of TF was carried out with the statistical package Genstat (Alvey, 1983) and is described by Frissel & Koster (1987). The geometric mean transfer factors were calculated as well as the 95% confidence interval with multiple regression analysis based on Eqs. 2 and 3. The amount of irrigation, which sometimes is an important variable, was neglected for the situation in the Netherlands. The uncertainty factor UF is calculated by UF = $[\exp(\sigma_{ln})]^2$ where σ_{ln} is the standard deviation of the log-transformed transfer data. UFs are based on selected sets of data. The following values of UF were found:
- not averaging time and space, UF = 2-12;
- one specific crop within a group of crops all belonging to one crop type, UF = 1.2-22;
- differences due to local conditions, UF = 4-10.

The expected contamination of crops

With the help of Table 1 and Eqs. 2 and 3 the expected contamination of the edible parts of crops can be estimated. For the Netherlands the total Cs-137 contamination varies between 3000 and 10000 Bq m^{-2}. With a uniform ditribution of this radioactivity over a 20-cm layer (10 cm in case of grass) the levels in soil vary from about 12-36 Bq kg^{-1}. For grassland soils these values are a factor of 2 higher. A calculation has been carried out for a contamination level of 5000 Bq m^{-2}, a clay soil with a pH

Table 3. Expected Cs-137 contamination of crops on a clay soil (for a specification of conditions see text).

Crop type[a]	Cs-137 contamination of crop products (Bq kg^{-1} dry weight)		
	expected value	lower limit	upper limit
Cereals	0.3	0.2	0.4
Fodder	0.9	0.6	1.3
Grass	4.3	3.0	5.7
Pods	1.5	0.9	2.3
Root crops	0.9	0.6	1.6
Tubers (potatoes)	1.3	0.9	2.0
Vegetables	3.4	2.3	4.8

[a] For specification see Table 2.

of 6, an average contamination time of 2 years and an organic matter content of 4% (10% for grassland). Results are shown in Table 3. It appears that the levels vary from 0.3 Bq kg^{-1} (dry weight base) for cereals to 4.3. Bq kg^{-1} for grass. The lower and upper 95% confidence limits are 0.2-0.4 for cereals and 3.0-5.7 for grass. It is emphasized that these values will be different for other conditions: a shift of the pH of one unit, for example, reduces or increases the uptake with a factor 2. It is mentioned again that these values apply only to values averaged for space and time.

Lysimeter experiments in the Netherlands

For a judgement of the validity of the derived equations within the Netherlands sufficient independently determined data are required. Such data are not available. Only RIVM has an ongoing programme on the uptake of radionuclides by grass, food and feed crops, and the data derived form a part of the data which were used for calculation of the Eqs. 2 and 3. Therefore, it is only possible to compare observed data in the Netherlands with the predicted values and to judge if these data agree. Disagreement might indicate a severe local effect, agreement the absence of such an effect.

Table 4. Comparison between measured and calculated transfer factors of radioactive Cs for crops grown on the RIVM lysimeter at Wageningen during 1986. Ratio = TF measured/TF calculated with Eq. 2 or 3. UF = uncertainty factor.

Crop	Loess soil			Sandy soil			Clay soil		
	number of samples	ratio	UF	number of samples	ratio	UF	number of samples	ratio	UF
Grass	6	0.60	3.4	6	3.4	4.0			
Beans	12	0.36	3.3	8	0.51	2.3	3	0.61	2.9
Spinach	12	3.8	3.5	8	2.0	1.8	4	3.9	2.0

The experiments at RIVM were carried out on a lysimeter which was installed in Wageningen in 1981. It contains 22 containers, most of which have a surface of 1 to 3 m^2; the depth is 1.4 m. At the bottom of the containers is an outlet from which percolated water is collected. Three soil types are used, a sandy soil (a humic podsol), a loess soil (an orthic luvisol) and a clay soil (an eutric fluvisol). Details are described by Stoutjesdijk et al., (1983). During the first four years a crop rotation of potatoes/cabbage - maize - beans/spinach - barley was used. Part of the lysimeter is used for permanent pasture. In 1986 the variation associated with individual containers was investigated. The results are shown in Table 4. This table shows the ratio between the observed TF values and the ones calculated with the Eqs. 2 or 3. The number of samples and the uncertainty factor calculated by the equation for UF, mentioned before, is also shown. Note that the ratio may be smaller or larger than 1, but that the UF is always larger than 1. Therefore an UF of 3.4 explains a ratio of 0.3 sufficiently. In general, it can be concluded from Table 4 that, for the circumstances tested, there is no indication of a severe deviation from the predicted uptake due to local conditions in the Netherlands. Therefore Eqs. 2 and 3 can be applied to estimate the crop uptake of Cs-137 in the Netherlands.

Final remark

This article has focussed all attention on Cs-134 and Cs-137 because these nuclides were the main long-lived radiocontaminants deposited after the Chernobyl accident. The studies at RIVM as well as the studies by the IUR take also into consideration other radionuclides such as strontium, plutonium, americium and neptunium.

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